

Comment on “Single Hole Dynamics in the CuO₂ Plane at Half Filling”

In a recent letter [1], Pothuizen *et al.* reported angle-resolved photoemission (ARPES) data on the single-layer insulating cuprate Sr₂CuO₂Cl₂. They observed a strongly \mathbf{k} dependent intensity: closest to ϵ_F , a weak feature near $(\pi/2, \pi/2)$ (assigned to the Zhang-Rice singlet state [2], consistent with Ref. [3]), and at high-symmetry points of the Brillouin zone and lower energy, relatively sharp peaks with large intensity. The spectra were analyzed in terms of a nearest neighbour (nn) tight-binding model using all O 2*p* and Cu 3*d* orbitals. From this, it was concluded that the strong intensity at the high-symmetry points comes from *purely oxygen-like* orbitals which due to the lack of 3*d* hybridization are very much single-particle-like. We argue that an assignment of the intense features to pure oxygen states is too simple, and inconsistent with LDA calculations for Sr₂CuO₂Cl₂ [4], which are expected to give accurate results for the single-particle properties we are interested in here.

An interpretation of the intense peaks in terms of pure oxygen states is *only* possible at the zone corner $\mathbf{k} = (\pi, \pi)$ for the state in Fig. 1c. It has *g* symmetry with respect to the Cu site, and therefore, cannot mix with any Cu 3*d* or Cl 3*p* levels. Furthermore, the LDA results [4] show that the admixture of Sr 3*d* levels is also negligible because of their large energy separation from the O 2*p* states.

However, the situation is entirely different at $\mathbf{k} = (0, 0)$ and $\mathbf{k} = (\pi, 0)$: In both cases, the O states used in the analysis of Ref. [1] can hybridize effectively with Cl 3*p_{x,y}* levels as shown in Figs. 1a,b. This is confirmed by LDA [4] which shows similar admixtures of O 2*p* and Cl 3*p* levels for the states closest to ϵ_F at those \mathbf{k} points. Consequently, it is not possible to extract a *generic* (usable as a microscopic model parameter for *all* cuprates) nn O-O hopping matrix element t_{pp} from the energy difference of the intense peaks at the different high-symmetry points as proposed in [1]. There are too many additional *material-specific* parameters (at least the O-Cl hopping matrix element, and the O-Cl on-site energy difference) which enter the expression for the energy splittings, not just t_{pp} .

Nevertheless, we agree with the authors of [1] that the strong intensity of these peaks is most likely due to the absence of mixing with the strongly correlated *d* states. This interpretation is further suggested by the fact that at $\mathbf{k} = (\pi, 0)$ the LDA predicts the topmost band below ϵ_F to have strong Cu 3*d_{x²-y²}* admixture, which might explain why the corresponding feature in the ARPES is not very sharp.

Another point concerns the width of the intense peaks: The LDA results show that at none of the \mathbf{k} points considered is there a *single* well separated band with no *d* character just below ϵ_F . There are always at least two bands with planar character, split by an amount comparable or less than 400meV, the approximate experimental width of the sharp peaks. Hence, their spectral weight is possibly a superposition from more than one band. This might also be partially responsible for their large intensity. Furthermore, a substantial

part of the line-width broadening is due to momentum resolution. The size of this contribution depends on the dispersion, and is therefore generally different at different \mathbf{k} points [5]. In any case, conclusions about scattering mechanisms from a comparison of line-widths as attempted in [1] have to be taken with caution.

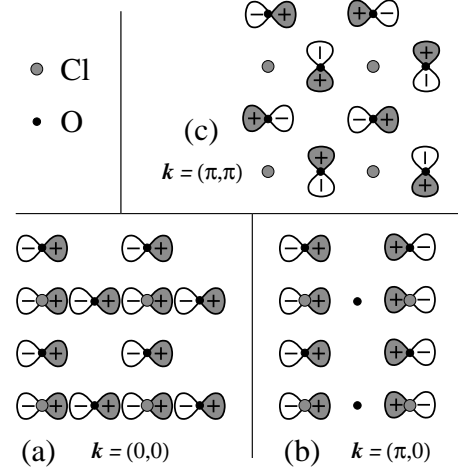


Fig. 1. Possible combinations of Oxygen 2*p* and Chlorine 3*p* orbitals at high-symmetry points of the Brillouin zone, (a) $\mathbf{k} = (0, 0)$, (b) $\mathbf{k} = (\pi, 0)$, (c) $\mathbf{k} = (\pi, \pi)$.

Finally, we would like to mention that LDA indeed predicts that the state closest to ϵ_F at $\mathbf{k} = (\pi, \pi)$ is the one depicted in Fig. 1c (almost degenerate with a mixed Cl 3*p_z*, Cu 3*d_{3z²-r²}* level), as proposed in [1]. The energy difference of this state as compared to the Zhang-Rice singlet feature is crucial in determining the stability of the Pr^{IV} oxidation state, which is thought to be responsible for the T_c suppression in PrBa₂Cu₃O_{7- δ} [6]. The experiments in [1] therefore impose some experimental constraints for the possible parameters of the model proposed in [6]. We shall elaborate on this point in future work.

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